Claims

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We claim:

1. A process for preparing 2,3,5,6-tetrafluorodimethylolbenzene, represented by the following structural formula, as one intermediate for pyrethroids,

$$F$$
 F
 CH_2OH

which comprising a step of reducing dialkyl 2,3,5,6-tetrafluoro terephthalate in the presence of a reductant and a solvent according to the following scheme:

FOOR₁

$$F \downarrow GOOR_2$$

$$F \downarrow GOOR$$

in the above formulae, R₁ and R₂ each independently represents a straight or branched alkyl chain having 1 to 6 carbon atoms, preferably, methyl, ethyl, n-propyl, n-butyl, n-pentyl, n-hexyl, iso-propyl, iso-butyl, tert-butyl or neopentyl.

- 2. The process according to claim 1, wherein both R₁ and R₂ represent methyl.
- 3. The process according to claim 1, wherein the solvent is selected from a group consisting of: alcohols; glycols; ethers; glycol ethers; glymes; polyglymes; polyethers; lower alcohols; two-phase solvent mixtures; polar inert solvents; organic acids; esters; water; ethers; lower anion surfactants or mixture thereof; or mixtures of two or more among them.
- 4. The process according to claim 3, wherein the solvent is methanol, ethanol, isopropanol, ethylene glycol, polyethylene glycols, diethyl ether, 1,2-dimethoxyethane, dioxane, tetrahydrofuran, diglyme or polyglyme, toluene, xylene, anisole, acetic acid, ethyl acetate, ethyl formate, water, tetrahydrofuran, or a mixture of two or more among them.
- 5. The process according to claim 3, wherein the solvent is methanol, ethanol, or a mixture thereof.

- 6. The process according to claim 1, wherein the reductant is a metal hydride, a borohydride, a metal aluminium hydride, aluminium borohydride, hydrogen or a hydrogen donor.
- 7. The process according to claim 6, wherein the borohydride is at least one selected from a group consisting of potassium borohydride, sodium borohydride, and lithium borohydride; and the metal aluminium hydride is lithium aluminium hydride.

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- 8. The process according to claim 1, wherein the reductant is potassium borohydride, sodium borohydride or lithium borohydride and the process is carried out in the presence of a catalyst or an accelerant.
- 9. The process according to claim 8, wherein the accelerant is a denaturated metal salt or a boride.
 - 10. The process according to claim 9, wherein the denaturated metal salt is one or more selected from a group consisting of aluminium, zinc, or titanium salts.
 - 11. The process according to claim 10, wherein the denaturated metal salt is one or more selected from a group consisting of aluminium chloride, zinc chloride, and titanium tetrachloride.
 - 12. The process according to claim 9, wherein the boride is boron trifluoride or alkyl boride.
 - 13. The process according to claim 8, wherein the accelerant is lithium compound, preferably lithium chloride or lithium bromide, when using potassium borohydride or sodium borohydride as a reductant.
- 14. The process according to claim 8, wherein the molar ratio of the accelerant to the reductant is 0.05-1:1, preferably, 0.1-0.5:1.
 - 15. The process according to claim 8, wherein the catalyst is onium salt, preferably one or more selected from a group consisting of tetra-alkyl ammonium salt, phosphonium salt, acyclic or cyclic-polyether.
- 16. The process according to claim 8, wherein the molar ratio of the catalyst to the reductant is 0.01-0.1:1.
 - 17. The process according to claim 4, wherein the solvent is methanol, ethanol, isopropanol, ethylene glycol, polyethylene glycol, diethyl ether, dioxane, tetrahydrofuran, 1,2-dimethoxyethane, diglyme or polyglyme.
- 18. The process according to claim 1, wherein the reduction is carried out using hydrogen as

the reductant in the presence of at least one catalyst selected from a group consisting of metals, metal oxides, mixed metal oxides, metal salts or metal complex catalysts.

- 19. The process according to claim 18, wherein the solvent is an alcohol, an aromatic hydrocarbon, an ether, an organic acid or ester thereof.
- 20. The process according to any one of preceding claims, wherein the process is carried out at from -20°C up to the boiling point of the solvent and is preferably carried out in the range of 30-120°C, more preferably in the range of 40-80°C.
 - 21. The process according to claim 1, wherein tefluthrin may readily be prepared by converting 2,3,5,6-tetrafluorodimethylolbenzene with the steps of halogenation, hydrogenation and esterification as follows:
 - i) halogenation of 2,3,5,6-tetrafluorodimethylolbenzene to give a 2,3,5,6-tetrafluoro-4-(halomethyl)benzyl alcohol;
 - ii) hydrogenation of the 2,3,5,6-tetrafluoro-4-(halomethyl)benzyl alcohol to give 4-methyl-2,3,5,6-tetrafluorobenzyl alcohol;
- 15 iii) esterification of 4-methyl-2,3,5,6-tetrafluorobenzyl alcohol with cis-((Z)-2-chloro-3,3,3-trifluoro-prop-1-enyl)-2,2-dimethylcyclopropane acyl chloride or cis-((Z)-2-chloro-3,3,3-trifluoro-prop-1-enyl)-2,2-dimethylcyclopropanecarboxylic acid to give tefluthrin.
 - 22. An intermediate compound of formula (II),

wherein,

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R₂ is a straight or branched alkyl chain having 1 to 6 carbon atoms and is preferably, methyl, ethyl, n-propyl, n-butyl, n-pentyl, n-hexyl, iso-propyl, iso-butyl, tert-butyl or neopentyl.

23. The compound according to claim 22, wherein R₂ is methyl.

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